PCT WORLD INTELLECTUAL PROPERTY ORGANIZATION
International Bureau

INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification 6 :
C10M 169/06 // (C10M 169/06, 115:08, 119:24, 135:18, 137:04), C10N 10:12, 40:00, 40:04, 50:10

A1

(11) International Publication Number: **WO 98/17748**

(43) International Publication Date: 30 April 1998 (30.04.98)

(21) International Application Number: PCT/EP97/05914

(22) International Filing Date: 17 October 1997 (17.10.97)

(30) Priority Data:
8/297278 18 October 1996 (18.10.96) JP

(71) Applicant *(for all designated States except US)*: SHELL INTERNATIONALE RESEARCH MAATSCHAPPIJ B.V. [NL/NL]; Carel van Bylandtlaan 30, NL–2596 HR The Hague (NL).

(72) Inventors; and
(75) Inventors/Applicants *(for US only)*: MASUMORI, Ryuichi [JP/JP]; (JP). OZAKI, Takahiro [JP/JP]; (JP). MUNAKATA, Tomoo [JP/JP]; (JP). YOSHIDA, Tsutomu [JP/JP]; Daiba Frontier Building, 3–2, Daiba 2–chome, Minato–ku, Tokyo 135 (JP).

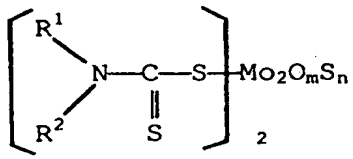
(81) Designated States: AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GE, GH, HU, ID, IL, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, US, UZ, VN, YU, ZW, ARIPO patent (GH, KE, LS, MW, SD, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, ML, MR, NE, SN, TD, TG).

Published
With international search report.
Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.

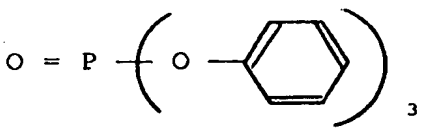
(54) Title: UREA GREASE COMPOSITION

(57) Abstract

A urea grease composition and gears containing such composition, which composition comprises a urea grease and as additives: (A) a molybdenum dialkyldithiocarbamate sulfide represented by formula (I), wherein R^1 and R^2 each independently represents a group selected from the group consisting of alkyl groups having from 1 to 24 carbon atoms; m is 0 or an integer of from 1 to 3; and n is an integer of from 1 to 4; provided that the sum of m and n is 4, and (B) a triphenyl phosphate represented by formula (II).



(I)



(II)

FOR THE PURPOSES OF INFORMATION ONLY

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AL	Albania	**ES**	Spain	**LS**	Lesotho	**SI**	Slovenia
AM	Armenia	**FI**	Finland	**LT**	Lithuania	**SK**	Slovakia
AT	Austria	**FR**	France	**LU**	Luxembourg	**SN**	Senegal
AU	Australia	**GA**	Gabon	**LV**	Latvia	**SZ**	Swaziland
AZ	Azerbaijan	**GB**	United Kingdom	**MC**	Monaco	**TD**	Chad
BA	Bosnia and Herzegovina	**GE**	Georgia	**MD**	Republic of Moldova	**TG**	Togo
BB	Barbados	**GH**	Ghana	**MG**	Madagascar	**TJ**	Tajikistan
BE	Belgium	**GN**	Guinea	**MK**	The former Yugoslav Republic of Macedonia	**TM**	Turkmenistan
BF	Burkina Faso	**GR**	Greece			**TR**	Turkey
BG	Bulgaria	**HU**	Hungary	**ML**	Mali	**TT**	Trinidad and Tobago
BJ	Benin	**IE**	Ireland	**MN**	Mongolia	**UA**	Ukraine
BR	Brazil	**IL**	Israel	**MR**	Mauritania	**UG**	Uganda
BY	Belarus	**IS**	Iceland	**MW**	Malawi	**US**	United States of America
CA	Canada	**IT**	Italy	**MX**	Mexico	**UZ**	Uzbekistan
CF	Central African Republic	**JP**	Japan	**NE**	Niger	**VN**	Viet Nam
CG	Congo	**KE**	Kenya	**NL**	Netherlands	**YU**	Yugoslavia
CH	Switzerland	**KG**	Kyrgyzstan	**NO**	Norway	**ZW**	Zimbabwe
CI	Côte d'Ivoire	**KP**	Democratic People's Republic of Korea	**NZ**	New Zealand		
CM	Cameroon			**PL**	Poland		
CN	China	**KR**	Republic of Korea	**PT**	Portugal		
CU	Cuba	**KZ**	Kazakstan	**RO**	Romania		
CZ	Czech Republic	**LC**	Saint Lucia	**RU**	Russian Federation		
DE	Germany	**LI**	Liechtenstein	**SD**	Sudan		
DK	Denmark	**LK**	Sri Lanka	**SE**	Sweden		
EE	Estonia	**LR**	Liberia	**SG**	Singapore		

UREA GREASE COMPOSITION

The present invention relates to urea grease compositions and to gears containing such compositions. Petroleum cost has increased sharply since so-called Oil Shock, and resourcing saving and energy saving ideas have been constantly intended in every industrial field, reflecting its influence.

In the motor vehicle industry, compact and light general motor vehicles have been developed, and the number of vehicles employing constant velocity joints (hereinafter, abbreviated as CVJ) have increased. Further, CVJ is also employed in 4 wheel drive vehicles and vehicles of four wheel independent suspension type, and the demand is growing. CVJ, which is also called constant velocity universal joint, is a joint which transmits rotation keeping constant angular speed and torque, and has different types depending on various uses. The lubricating agents used for the CVJ are exposed to severer condition as a motor vehicle achieves higher power and higher speed and CVJ itself becomes compact, and therefore, a grease is demanded which reduces friction and wear on sliding portions. Further, a seal boot member is required to be mounted on CVJ to prevent leaking of a grease and to prevent invasion of foreign materials and water from outside. As for this material, chloroprene rubber is generally used and polyester resins are also commonly used.

A grease which is excellent in low friction and wear resistance suppresses vibration and noise of a vehicle body at a time of starting and accelerating and during driving. Moreover, due to its temperature suppressing

effect, durability of a seal boot member can be improved. However, excessive increase of temperature promotes ageing of the seal boot member and degradation of a lubricating agent and extremely shortens the life of CVJ.

On the other hand, in the steel industry, continuous operation of mechanical equipment has been further developed. With the technical revolution, there have been a strong demand in high quality of a product, improvement in production process capacity, ensuring of reliability of an apparatus, and the like. Lubrication conditions are very severe, since greases used in these mechanical apparatus are in a tough environment, that is, high temperature, contact with water, high possibility of invasion of foreign materials such as scale, and the like, and are continuously exposed to severe conditions day and night. A grease is required which has excellent seal resistance and reduces friction and wear to lengthen the life of mechanical parts and to prevent sudden failure to the utmost. Further, the parts to be lubricated of industrial machinery, machine tools, and the like suffer from considerable friction, and excessive wear causes reduction in accuracy of the machine, so that the mechanical parts have to be replaced. As described above, to reduce friction and save wear is an important object of the present invention.

Therefore, in the parts to be lubricated as described above, lithium-based greases have been conveniently used using sulfur-based compounds such as sulfurized fat and oil, sulfurized olefin and the like, lead naphthenate, metal dithiophosphate, metal dithiocarbamate-based additive and the like. Recently,

a lithium complex grease and a urea grease which are superior in heat resistance to lithium-based greases have been increasingly used.

Under this situation, as one of typical prior art, U.S. Patent No. 4,514,312 discloses a grease using aromatic amine phosphate as an organic additive containing no sulfur in a urea grease. Further, U.S. Patent No. 4,840,740 discloses a grease comprising a urea gresae, an organic molybdenum compound, and zinc dithiophosphate. Japanese Patent Publication (JP-B) No. 04-34590(1992) discloses a urea grease comprising as an essential component a sulfur-phosphorus-based extreme pressure additive comprising

1) molybdenum dialkyldithiocarbamate sulfide and
2) one or a combination of two or more selected from the group consisting of sulfurized fat and oil, sulfurized olefin, tricresyl phosphate, trialkyl thiophosphate and zinc dialkyl dithiophosphate.

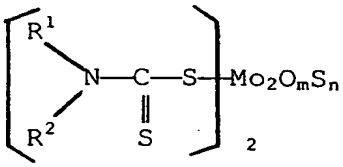
Although some of the greases in the prior art are recognized to have reducing effects in friction and wear, full satisfaction has not been attained. Further, there are disadvantages that sealants deteriorate under high temperature. For example, an aromatic amine phosphate and lead naphthenate degrade polyester resins and a sulfurized fat and oil degrades chloroprene rubber, respectively, and a sulfurized olefin extremely degrades both chloroprene rubber and polyester resins.

Further, in other prior art, Japanese Patent Application Laid-Open (JP-A) No. 08-157859 (1996) discloses the use of as additives a molybdenum dialkyldithiocarbamate sulfide and triphenylphosphorothionate, and Japanese Patent Application Laid-Open (JP-A) No. 62-2275197 (1987) discloses the use of as

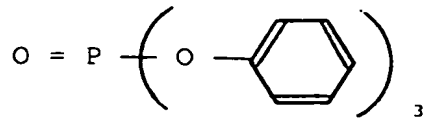
thickening agents a lithium soap of 12-hydroxy fatty acid, lithium phosphate formed from a phosphate such as trimethyl phosphate or triphenyl phosphate. Further, Japanese Patent Application Laid-Open (JP-A) No. 03-231993 (1991) discloses a grease composition using a phosphate oil in a urea compound.

The present invention relates to a urea grease which is excellent in frictional properties and wear resistance and also has good fitting property to sealants such as chloroprene rubber and polyester resin. More particularly, the present invention relates to a urea grease composition suitable for application to such parts to be grease-lubricated as constant velocity joints, ball joints, and ball bearings in motor vehicles and bearings and gears of various industrial equipments such as the steel and industrial machines and machine tools.

The present invention relates to a urea grease composition comprising a urea grease and as additives (A) a molybdenum dialkyldithiocarbamate sulfide represented by the formula:



wherein R^1 and R^2 each independently represents a group selected from the group consisting of alkyl groups having from 1 to 24 carbon atoms; m is 0 or an integer of from 1 to 3; and n is an integer of from 1 to 4; provided that the sum of m and n is 4, and (B) a triphenyl phosphate represented by the formula:



The compounds corresponding to the above-mentioned component (A) have a high melting point. Examples of those compounds include diethyl sulfide-molybdenum dithiocarbamate, dibutyl-molybdenum dithiocarbamate, diisobutyl sulfide-molybdenum dithiocarbamate, di(2-ethylhexyl)sulfide-molybdenum dithiocarbamate, diamyl sulfide-molybdenum dithiocarbamate, diisoamyl sulfide-molybdenum dithiocarbamate, dilauryl sulfide-molybdenum dithiocarbamate, distearyl sulfide-molybdenum dithiocarbamate. They are preferably mixed in the form of powder, and the amount added is from 0.5 to 10% by weight, preferably from 0.5 to 5% by weight based on the total weight. When the amount added is less than 0.5% by weight, effect of improvement in friction resistance and wear resistance is generally insufficient, and when over 10% by weight, no further improvement will generally occur.

As triphenyl phosphate has a melting point of 50°C and is solid at ambient temperature, it is preferred that the powder is blended with the base grease at a temperature of not less than 50°C. The amount of component (B) preferably is from 0.1 to 10% by weight, more preferably from 0.1 to 5% by weight based on the total weight. When the amount used is less than 0.1% by weight, improvement in friction resistance and wear resistance properties generally cannot be achieved, and when over 10% by weight, further lubricating ability generally cannot be obtained.

As to the urea compound, any of known thickening agents of urea type can be used. For example, diurea, triurea and tetraurea may be used.

As the base oil, a mineral oil and/or a synthetic oil can be used. If the urea compound is used as a thickening agent it is preferably applied in an amount from 2 to 35% by weight based on the total weight of the base oil and the urea compound.

Also, various additives such as an antioxidant, anticorrosion agent, extreme pressure agent, polymer and the like can further be added to the composition of the present invention.

The urea grease composition according to the invention are especially suitable for use in gears, more specifically constant velocity joints. Therefore, the present invention further relates to gears, more specifically constant velocity joints, containing such grease.

Example and Comparative Example

The present invention will be specifically described by Examples and Comparative Examples hereinafter, however it is not construed to be limited to them. Additives were added to base greases in blending ratios as shown in Tables 1 to 5 (all of them are shown in % by weight), the mixtures were treated by three-roll mill to obtain greases of Examples and Comparative Examples.

The formulations of the base greases are as described below. As the base oil, a purified mineral oil having a viscosity of 15 mm^2/sec at 100°C was used.

I. Diurea grease

One mol of diphenylmethane-4,4'-diisocyanate and 2 mol of octylamine were reacted in the base oil, and the produced urea compound was dispersed uniformly to obtain

a grease. The amount of urea compound was 10% by weight based on the total weight of the base oil and the urea compound.

II. Tetraurea grease

Two mol of diphenylmethane-4,4'-diisocyanate, 2 mol of octylamine, and 1 mol of ethylenediamine were reacted in the base oil, and the produced urea compound was dispersed uniformly to obtain a grease. The amount of urea compound was 15% by weight based on the total weight of the base oil and the urea compound.

III. Lithium complex grease

Hydrogenated caster oil fatty acid was dissolved in the base oil, to which was added an aqueous lithium hydroxide solution for neutralization, and the mixture was dehydrated during the reaction. After completion of the dehydration, azelaic acid was added. The reaction was carried out with the help of an aqueous lithium hydroxide solution in the amount necessary for neutralization. The lithium azelate and lithium 12-hydroxystearate soap were mixed and dispersed uniformly to obtain a grease. The amount of lithium 12-hydroxy-stearate as a thickening agent was 7.5% by weight and the amount of lithiumazelate was 2.5% by weight, respectively, based on the total weight of the base oil and the thickening agent.

The following test were conducted regarding friction coefficient, wear resistance and fitting property to a sealant as shown in the tables, and evaluations were made.

(1) Friction coefficient

Friction coefficient after 15 minutes was measured under the following conditions using a Falex testing machine (according to IP/241/69).

Rotation speed	:	290 rpm
Load	:	890N (200 lb)
Temperature	:	room temperature
Time	:	15 minutes
Amount of grease	:	about 1 g of grease was applied on a test piece

(2) Wear resistance

According to the four ball type wearing test of ASTM D2226

Rotation speed	:	1200 rpm
Load	:	392N (40 kgf)
Temperature	:	75°C
Time	:	60 minutes

(3) Fitting property to sealant

Sealants which were chloroprene rubber of a polyester resin were immersed in respective grease compositions and tested under the following conditions according to the vulcanized rubber physical test of JIS K6301. The tensile strength and elongation before and after the test were measured and change (%) was obtained.

Temperature	:	140°C
Immersion Time	:	96 hrs

Table 1

			Example			
			1	2	3	4
Formulation (% by weight)	Base grease	Diurea grease Tetraurea grease	96.5	94.5	96.0	94.0
	Additive	A-1[*1]	3.0		3.0	5.0
		A-2[*2]		5.0		
		B[*3]	0.5	0.5	1.0	1.0
	Total		100.00	100.00	100.00	100.00
Test result	Friction coefficient (μ)		0.092	0.089	0.090	0.084
	Wear resistance (mm)		0.42	0.40	0.41	0.41
	Chloroprene rubber	Change in Tensile strength (%)	-6.4	-5.3	+1.3	-5.3
		Change in Elongation (%)	-2.9	+0.9	+18.7	+5.2
	Polyester resin	Change in Tensile strength (%)	+2.5	-4.5	-20.7	-6.8
		Change in Elongation (%)	+14.0	-7.8	-15.1	-11.4

[*1]: A-1 is a molybdenum dialkyldithiocarbamate sulfide, MOLYVANA A, available from Vanderbilt Co..

[*2]: A-2 is a molybdenum dialkyldithiocarbamate sulfide, SAKURALUBE 600, available from Asahi Denka Kogyo K.K..

[*3]: B is a triphenyl phosphate.

Table 2

			Example		
			5	6	7
Formulation (% by weight)	Base grease	Diurea grease Tetraurea grease	95.0	96.0	93.0
	Additive	A-1*1 A-2*2 B*3	3.0 2.0	2.0 1.0 1.0	2.0 3.0 2.0
	Total		100.00	100.00	100.00
Test result	Friction coefficient (μ) Wear resistance (mm)		0.083 0.41	0.088 0.42	0.082 0.38
	Chloroprene rubber	Change in Tensile strength (%) Change in Elongation (%)	-6.3 +5.6	-8.1 +5.2	-3.9 +4.1
	Polyester resin	Change in Tensile strength (%) Change in Elongation (%)	-14.6 -1.9	-16.5 -10.6	-4.8 -5.9

*1: A-1 is a molybdenum dialkyldithiocarbamate sulfide, MOLYVANA A, available from Vanderbilt Co..

*2: A-2 is a molybdenum dialkyldithiocarbamate sulfide, SAKURALUBE 600, available from Asahi Denka Kogyo K.K..

*3: B is a triphenyl phosphate.

Table 3

			Comparative Example				
			1	2	3	4	5
Formulation (% by weight)	Base grease	Diurea grease Tetraurea grease Lithium complex grease	99.5	98.0	97.0	98.0	95.0
	Additive	A-1[*1] A-2[*2] B[*3] Vanlube 592[*4] Lubrizol 1360[*5] Lubrizol 5006[*6] Lubrizol 5340L[*7] Tricresyl phosphate Dailube L-30[*8]	0.5	2.0	3.0	2.0	3.0 2.0
	Total		100.00	100.00	100.00	100.00	100.00

[*1]: A-1 is a molybdenum dialkyldithiocarbamate sulfide, MOLYVAN A, available from Vanderbilt Co.
[*2]: A-2 is a molybdenum dialkyldithiocarbamate sulfide, SAKURALUBE 600, available from Asahi Denka Kogyo K.K..
[*3]: B is a triphenyl phosphate.
[*4]: Vanlube 592 is a trade name of R.T. Vanderbilt Co. Inc., and a viscous liquid (190 SUS/100°C) comprising an aromaticaminephosphate.
[*5]: Lubrizol 1360 is a zinc dialkyldithiophosphate.
[*6]: Lubrizol 5006 is a sulfurized fat and oil.
[*7]: Lubrizol 5340L is a sulfurized olefin.
[*8]: Dailube L-30 is a lead naphthenate of Dainippon Ink and Chemicals, Inc.

Table 4

			Comparative Example				
			6	7	8	9	10
Formulation (% by weight)	Base grease	Diurea grease Tetraurea grease Lithium complex grease	95.5	94.0	95.0	96.5	95.0
	Additive	A-1[*1] A-2[*2] B[*3] Vanlube 592[*4] Lubrizol 1360[*5] Lubrizol 5006[*6] Lubrizol 5340L[*7] Tricresyl phosphate Dailube L-30[*8]	3.0 1.0 0.5	3.0 1.0 2.0	3.0 2.0	3.0 0.5	3.0 2.0
	Total		100.00	100.00	100.00	100.00	100.00

[*1]: A-1 is a molybdenum dialkyldithiocarbamate sulfide, MOLYVAN A, available from Vanderbilt Co.
[*2]: A-2 is a molybdenum dialkyldithiocarbamate sulfide, SAKURALUBE 600, available from Asahi Denka Kogyo K.K..
[*3]: B is a triphenyl phosphate.
[*4]: Vanlube 592 is a trade name of R.T. Vanderbilt Co. Inc., and a viscous liquid (190 SUS/100°C) comprising an aromaticaminephosphate.
[*5]: Lubrizol 1360 is a zinc dialkyldithiophosphate.
[*6]: Lubrizol 5006 is a sulfurized fat and oil.
[*7]: Lubrizol 5340L is a sulfurized olefin.
[*8]: Dailube L-30 is a lead naphthenate of Dainippon Ink and Chemicals, Inc.

Table 5

			Comparative Example				
			1	2	3	4	5
Test result	Friction coefficient (μ)		0.144	0.126	0.116	0.120	0.103
	Wear resistance (mm)		0.56	0.58	0.44	0.51	0.41
	Chloroprene rubber	Change in Tensile Strength (%)	-5.3	-12.1	-18.4	-9.4	-16.9
		Change in Elongation (%)	-0.4	-4.1	-2.2	+29.5	+34.1
	Polyester resin	Change in Tensile strength (%)	-1.9	-14.2	-4.9	-36.0	+2.7
		Change in Elongation (%)	-3.2	+8.8	-0.5	-38.6	+34.3

Table 6

			Comparative Example				
			6	7	8	9	10
Test result	Friction coefficient (μ)		0.123	0.119	0.123	0.119	0.117
	Wear resistance (mm)		0.41	0.42	0.44	0.40	0.41
	Chloroprene rubber	Change in Tensile Strength (%)	-53.0	-74.8	-13.2	-10.0	-16.6
		Change in Elongation (%)	-31.5	-81.3	+23.9	+0.8	+7.6
	Polyester resin	Change in Tensile strength (%)	-17.7	-52.7	-43.2	+9.0	+7.2
		Change in Elongation (%)	-8.2	-54.0	-70.6	+23.3	+28.8

The friction coefficients in Examples 1 to 7 are all smaller as compared with those of Comparative Examples 1 to 10. Regarding wear resistance, Comparative Examples 5, 6, 7, 9 and 10 provide the same degree of good results as the examples according to the invention. However, these Comparative Examples are inferior in compatability to chloroprene rubber and/or polyester resin.

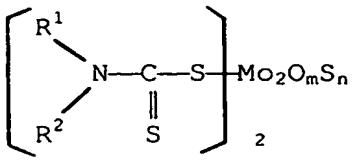
For example, Comparative Example 5 gives a large change in elongation of chloroprene rubber and polyester resin. Comparative Example 6 gives a large change in tensile strength and elongation of chloroprene rubber and polyester resin. Comparative Examples 9 and 10 give relatively large change in elongation of a polyester resin. Examples 1 to 7 give an excellent friction coefficient and wear resistance and also an excellent fitting property to sealants of chloroprene rubber and polyester resins.

The grease of the present invention has a low friction coefficient and is excellent in wear resistance. As a result, vibration and noise of CVJ of motor vehicles are suppressed. Further, the grease of the present invention reduces friction in ball joints and wheel bearings of motor vehicles and other various industry machine apparatuses, and can lengthen the life of machine parts.

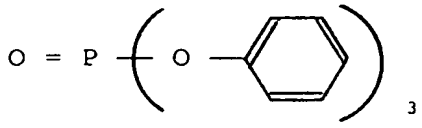
The grease of the present invention is excellent in the fitting property to chloroprene rubber and polyester resin, and delays the degradation of the sealant of a sealed apparatus even at high temperature.

C L A I M S

1. A urea grease composition comprising a urea grease and as additives (A) a molybdenum dialkyldithiocarbamate sulfide represented by the formula:



wherein R^1 and R^2 each independently represents a group selected from the group consisting of alkyl groups having from 1 to 24 carbon atoms; m is 0 or an integer of from 1 to 3; and n is an integer of from 1 to 4; provided that the sum of m and n is 4, and (B) a triphenyl phosphate represented by the formula:



2. The urea grease composition according to claim 1, wherein said component (A) and said component (B) are blended in an amount of from 0.5 to 10% by weight and from 0.1 to 10% by weight, respectively, based on the total weight of the urea grease composition.

3. The urea grease composition according to claim 1 or 2, wherein said urea grease is composed of a base oil and as a thickening agent a urea compound, the proportion of the urea compound being from 2 to 35% by weight based on the total weight of the base oil and the urea compound.

4. Gears containing a urea grease composition according to anyone of claims 1-3.

5. Gears according to claim 4, which gears are constant velocity joints.

INTERNATIONAL SEARCH REPORT

Inter nal Application No
PCT/EP 97/05914

A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 C10M169/06 //(C10M169/06,115:08,119:24,135:18,137:04), C10N10:12,40:00,40:04,50:10

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 6 C10M

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	DATABASE WPI Section Ch, Week 8742 Derwent Publications Ltd., London, GB; Class E19, AN 87-295595 XP002034609 & JP 62 207 397 A (KYODO YUSHI KK) , 11 September 1987 cited in the application	1-5
Y	see abstract	1-5

Y	EP 0 633 304 A (SHOWA SHELL SEKIYU; TOYOTA MOTOR CO LTD) 11 January 1995 see page 3, line 57 - page 4, line 5; tables 1,2	1-5

A	US 5 133 888 A (WAYNICK JOHN A) 28 July 1992 see claims 1,2; examples 2-6	1-3

☐ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

* Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

1

Date of the actual completion of the international search	Date of mailing of the international search report
19 February 1998	13. 03. 98

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Kazemi, P

Form PCT/ISA/210 (second sheet) (July 1992)

INTERNATIONAL SEARCH REPORT

information on patent family members

Inten nal Application No
PCT/EP 97/05914

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
EP 0633304 A	11-01-95	JP 6330072 A	29-11-94
		BR 9402060 A	27-12-94
		CA 2124283 A	26-11-94
		CN 1097796 A	25-01-95
		KR 9707496 B	09-05-97
		US 5449471 A	12-09-95
US 5133888 A	28-07-92	NONE	